Nonlinear magneto-optical diffraction from periodic domain structures in magnetic films

N. N. Dadoenkova
Donetsk Physical-Technical Institute of the National Academy of Sciences of Ukraine, 340114 Donetsk, Ukraine

I. L. Lyubchanskii
Donetsk Physical-Technical Institute of the National Academy of Sciences of Ukraine, 340114 Donetsk, Ukraine and Research Institute for Materials, University of Nijmegen, 6525 ED Nijmegen, The Netherlands

M. I. Lyubchanskii
Donetsk Physical-Technical Institute of the National Academy of Sciences of Ukraine, 340114 Donetsk, Ukraine

Th. Rasing[a]
Research Institute for Materials, University of Nijmegen, 6525 ED Nijmegen, The Netherlands

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The nonlinear optical diffraction in magnetic films with a laminar domain structure and Bloch-type domain walls is investigated for both *s* and *p* polarization of incident light. It is shown that the contribution of magnetic domains and domain walls to the nonlinear diffraction can be separated by a polarization analysis of the scattered light.
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In the last few years new nonlinear optical methods have been explored to investigate magnetic films and multilayers.[1–4] Recently the observation of labyrinthlike magnetic domain structures in chromium oxide Cr_2O_3,[5] yttrium–iron garnets (YIG) $Y_3Fe_5O_{12}$,[6] and yttrium–manganese–oxide $YMnO_3$[7] via magnetically induced optical second harmonic generation (MSHG) was reported. Results of the theoretical investigations of MSHG on magnetic domains (MDs) and domain walls (DWs) were published in Refs. 8–10.

It is well known that laminar (one-dimensional) periodic domain structures appear in magnetic thin films under the influence of an internal magnetic field $\mathbf{H}_0$ directed perpendicularly to the film. Because the magnetic domain sizes are comparable to the wavelengths of the fundamental and second harmonic light one should expect the appearance of linear and nonlinear diffraction from such a domain structure. Generally speaking, a periodic domain structure can be presented as a diffraction grating which modulates the linear as well as the nonlinear magneto-optical susceptibilities. As a result, MSHG will be sensitive to the existence of a periodic domain structure and nonlinear magneto-optical diffraction (at the second harmonic frequency) can arise. The linear diffraction from laminar magnetic domain structures was investigated in numerous publications (see, for example, Ref. 11 and the monograph in Ref. 12), but so far, the nonlinear diffraction has not been considered. On the other hand, SHG and nonlinear diffraction in ferroelectric films with a laminar domain structure, needlelike ferroelectric domains and periodically poled ferroelectrics were studied both theoretically and experimentally starting from 1968[13] (see also Refs. 14 and 15, and some recent publications[16–20]). Recently the second harmonic imaging of ferroelectric DWs was reported.[21–23]

Nonlinear magneto-optical investigations of magnetic films and structures have several advantages in comparison to their linear equivalents. First, the nonlinear magneto-optical response allows us to obtain information about the magnetization at surfaces and buried interfaces.[3] Second, as was shown in experiments with YIG films,[6] with nonlinear magneto-optics it is possible to observe peculiarities of domain structures which are absent in the usual linear optical response. Third, nonlinear magneto-optics yields totally new effects like the observation of antiferromagnetic domains[5] and a transversal nonlinear magneto-optical effect linear in the magnetization.[24] Therefore, it should be expected that nonlinear magneto-optical diffraction will allow us to get more detailed information about periodic domain structures as well as about contributions of the domain walls to the nonlinear magneto-optical response.

The aim of this letter is to show the possibility of the observation of the nonlinear magneto-optical diffraction in magnetic films with a laminar periodic magnetic domain structure.

Let us consider the following geometry: a laminar domain structure with Bloch-type DWs is located in the *XY* plane and the *Z* axis is perpendicular to the film. The domains are then oriented along the *X* axis and form a regular structure along the *Y* axis with a period *D*

$$D = d_+ + d_- + 2d_{\mathrm{DW}}, \tag{1}$$

where d_+ and d_- are the widths of the MDs with reversed magnetization directions and d_{DW} is the width of the DWs,

[a]Electronic mail: rim@sci.kun.nl

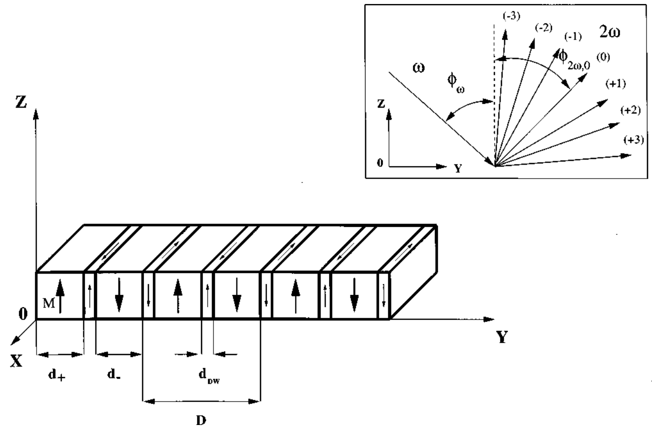

FIG. 1. Schematic image of magnetic film with a laminar domain structure. Arrows in magnetic domains and domain walls indicate the orientation of magnetization. Directions of incident (at the frequency ω) and diffracted (at the frequency 2ω) light are shown in inset.

respectively (see Fig. 1). The magnetization M in the domains is directed along the Z axis and in the DWs it is oriented in the XZ plane.

The nonlinear polarization $\mathbf{P}^{\mathrm{NL}}(2\omega)$ at the frequency 2ω in the dipole approximation can be presented as[25]

$$P_i^{\mathrm{NL}}(2\omega)=\chi_{ijk}(-2\omega:\omega,\omega)E_j(\omega)E_k(\omega), \tag{2}$$

where $\mathbf{E}(\omega)$ is the electric field of the incident light at the frequency ω and χ_{ijk} is the quadratic nonlinear optical susceptibility tensor, which has for a magnetic film the following form:[8,9]

$$\chi_{ijk}=\chi_{ijk}^{(0)}+\chi_{ijk}^{(m)}. \tag{3}$$

Here $\chi_{ijk}^{(0)}$ and $\chi_{ijk}^{(m)}$ are the magnetic ordering independent and dependent parts of χ_{ijk}, respectively. Following Refs. 8 and 9, the magnetic ordering induced nonlinear optical susceptibility tensor $\chi_{ijk}^{(m)}$ can be presented as an expansion in the magnetization and magnetization gradients

$$\chi_{ijk}^{(m)}=\chi_{ijkL}^{(m,1)}M_L+\chi_{ijkLM}^{(m,2)}M_LM_M+\chi_{ijklM}^{(m,3)}\frac{dM_M}{dr_l}+\chi_{ijkLmN}^{(m,4)}M_L\frac{dM_N}{dr_m}+\dots, \tag{4}$$

where $\hat{\chi}^{(m,\alpha)}$ are the nonlinear magneto-optical tensors.[8,9]

Because in our case the MDs in a magnetic film are ordered in one direction (along the Y axis), the magnetization induced nonlinear optical susceptibility tensor $\chi_{ijk}^{(m)}$ in such a laminar structure is sensitive to the magnetization distribution in the film, and can be presented in the following form:

$$\chi_{ijk}^{(m)}=\sum_{n_y}\chi_{ijk}^{(m)}(n_y)\exp(iQn_yy), \tag{5}$$

where $Q=2\pi/D$ is the reciprocal vector of the laminar magnetic domain structure and n_y is an integer.

The directions to observe diffracted second harmonic light will be determined by

$$k_{2\omega}\sin\phi_{2\omega,N}=2k_\omega\sin\phi_\omega+NQ, \tag{6}$$

where $k_{2\omega}$ and k_ω are the wave numbers of second harmonic and incident light, ϕ_ω and $\phi_{2\omega,N}$ are the angle of incidence and the angle of the Nth order second harmonic diffraction, respectively. Equation (6) is the nonlinear analog of Bragg's law for a three-wave interaction.[14,16] From Eq. (6) it follows that the diffraction order N can be determined by the relation

$$N<\frac{2D}{\lambda_\omega}\left(\frac{n_{2\omega}}{n_\omega}-\sin\phi_\omega\right), \tag{7}$$

where n_ω and $n_{2\omega}$ are the refractive indices at the fundamental and second harmonic frequencies.

For YIG films, which were studied in Ref. 6, the average sizes of MDs and DWs were equal to

$$d_+\simeq d_-\simeq 2\ \mu\mathrm{m},\quad d_{\mathrm{DW}}\simeq 0.1\ \mu\mathrm{m}. \tag{8}$$

For an incident light beam with $\lambda_\omega=0.775\ \mu$m (Ti:sapphire laser) and $\phi_\omega=45°$, we conclude from Eq. (7) that it would be possible to observe three nonlinear diffraction orders at the following angles (see Fig. 1):

$$\phi_{2\omega,3}=79.67°,\quad \phi_{2\omega,-3}=25.48°,\quad \phi_{2\omega,2}=63.06°,$$

$$\phi_{2\omega,-2}=31.50°,\quad \phi_{2\omega,1}=53.06°,\quad \phi_{2\omega,-1}=37.93°.$$

Using our results of Ref. 9, we can now find the contribution from the different terms in Eq. (4) to the nonlinear polarization for s and p out polarizations.

(1) S-polarized incident light, i.e., $\mathbf{E}(\omega)=[E_x(\omega),0,0]$:

$$P_x^{\mathrm{NL}}(2\omega)=\chi_{xxxyZ}^{(m,3)}\frac{dM_Z}{dy}E_x^2(\omega), \tag{9}$$

$$P_y^{\mathrm{NL}}(2\omega)=\chi_{yxxZ}^{(m,1)}M_ZE_x^2(\omega), \tag{10}$$

$$P_z^{\mathrm{NL}}(2\omega)=(\chi_{zxx}^{(0)}+\chi_{zxxXX}^{(m,2)}M_X^2+\chi_{zxxZZ}^{(m,2)}M_Z^2)E_x^2(\omega). \tag{11}$$

Equation (9) corresponds to the s-polarized nonlinear polarization and describes the contribution of DWs (via the magnetization gradient), whereas Eqs. (10) and (11) give the p-polarized output. However, the p-polarized nonlinear polarization is determined by a nonmagnetic contribution [first term in Eq. (11)] and by both MDs (terms linear and quadratic in M_Z) and DWs (all magnetically dependent terms, because the magnetization vector in Bloch-type DWs contains M_X and M_Z components).

(2) P-polarized incident light, i.e., $\mathbf{E}(\omega)=[0,E_y(\omega),E_z(\omega)]$:

$$P_x^{\mathrm{NL}}(2\omega)=\left[\chi_{xyzZ}^{(m,1)}M_Z+2\left(\chi_{xyzXyZ}^{(m,4)}M_X\frac{dM_Z}{dy}+\chi_{xyzZyX}^{(m,4)}M_Z\frac{dM_X}{dy}\right)\right]E_y(\omega)E_z(\omega), \tag{12}$$

$$P_y^{\mathrm{NL}}(2\omega)=(\chi_{yyz}^{(0)}+\chi_{yyzXX}^{(m,2)}M_X^2+\chi_{yyzZZ}^{(m,2)}M_Z^2)E_y(\omega)E_z(\omega)-\chi_{yyyX}^{(m,1)}M_XE_y^2(\omega)-\chi_{yzzX}^{(m,1)}M_XE_z^2(\omega), \tag{13}$$

$$P_z^{\mathrm{NL}}(2\omega)=(\chi_{zyy}^{(0)}+\chi_{zyyXX}^{(m,2)}M_X^2+\chi_{zyyZZ}^{(m,2)}M_Z^2)E_y^2(\omega)+(\chi_{zzz}^{(0)}+\chi_{zzzXX}^{(m,2)}M_X^2+\chi_{zzzZZ}^{(m,2)}M_Z^2)E_z^2(\omega)-\chi_{zyzX}^{(m,1)}M_XE_y(\omega)E_z(\omega). \tag{14}$$

In contrast to Eq. (9), it follows from Eq. (12) that not only DWs, but also MDs will contribute (via term proportional to M_Z) to the s-polarized second harmonic radiation for p-polarized incident light. This means that for the experimental geometry $s(\omega)\rightarrow s(2\omega)$ it should be possible to detect diffracted second harmonic radiation which is induced by Bloch-type DWs only.

Because the widths of MDs and the period of a laminar domain structure are very sensitive to an external magnetic field $\mathbf{H}_0$, there exists the possibility to magnetically control the diffracted second harmonic radiation. As was shown in Ref. 26, at $H_0 > 0.4\pi M_s$ (M_s is the saturation magnetization), the period D and the width of the positive domain d_+ (in which the magnetization is oriented parallel to $\mathbf{H}_0$) rapidly increase nonlinearly (approximately quadratically) with H_0, whereas the width of the negative domain d_- decreases much more slowly (though also nonlinearly). Thus, the angles which determine the nonlinear diffraction maxima $\phi_{2\omega,N}$ in Eq. (6) will change as well. For $H_0 > 4\pi M_s$ the magnetic film will transit to the uniform magnetic state and for the geometry $s(\omega)\rightarrow s(2\omega)$ second harmonic radiation will disappear.

We would like to note that for ferroelectrics the situation is very similar. Recently, an analysis of the selection rules for the nonlinear polarization in ferroelectric crystals with DWs was made in Ref. 22. However, the authors of this article only took into account the nonlinear susceptibility tensor components which correspond to the domains. For a complete description of the DW contributions to the SHG signal in nonuniform ferroelectrics, it is necessary to also take into account additional terms in the nonlinear optical susceptibility tensor that depend on the electric polarization as well as the polarization gradient. In the same way as was shown above for the magnetic domain structure, in ferroelectric media the gradient terms of the electric polarization determine the DWs contribution to the formation of second harmonic radiation. It can be shown that for ferroelectric crystals of T_d symmetry the polarization gradient terms will also lead to a nonlinear polarization at the second harmonic frequency for $s(\omega)\rightarrow s(2\omega)$ and $p(\omega)\rightarrow s(2\omega)$ geometries[27] and can thus be distinguished from the domain contributions. The first nonzero terms that give a DW contribution are given by the following terms in nonlinear optical susceptibility tensor: $\chi_{ijk} \sim \chi_{ijkzy}^{(e,3)}(dP_z/dy)$ where $\chi_{ijkzy}^{(e,3)}$ is the corresponding tensor coefficient in the expansion of χ_{ijk} on components of the polarization $\mathbf{P}$ and the polarization gradient.[27] The fact that domain boundaries are characterized by different (than domains) nonlinear optical susceptibility tensor components was recently realized in Ref. 23, however, without giving any explicit derivations.

In conclusion, in this letter we showed that a nonlinear magneto-optical investigation of ordered magnetic structures is very attractive, because it allows us to separate contributions of magnetic domains and domain walls via polarization measurements. In addition, the treatment above can be extended to two-dimensional ordered magnetic domains, like magnetic bubble lattices[28] or biperiodic stripe domain structures, which were observed in magnetic garnet films recently.[29]

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